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Bound exciton states of isoelectronic centers in GaAs:N grown by an atomically controlled doping technique

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We have studied bound exciton states of isoelectronic centers of nitrogen-doped GaAs by photoluminescence (PL) spectroscopy. The nitrogen doping has been performed in an atomically controlled way using the (3×3) nitrogen stable surface on GaAs(001), which has succeeded in forming a series of distinct, strong, narrow bandwidth PL lines. The PL lines have been attributed to recombinations of excitons bound to nitrogen pairs ordered along $[110]$. Magneto PL demonstrates the Zeeman-level diagrams of the triplet state influenced by the defect anisotropy. The PL features of GaAs:N have been compared with the results for $\text{In}_x\text{Ga}_{1-x}\text{As:N}$

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I. INTRODUCTION

Nitrogen (N) has a strong electronegativity and creates isoelectronic centers in III-V alloy semiconductors. The incorporation of a small amount of N leads to significant changes in the local electronic structure of the host semiconductor. N-containing GaAs ($\text{GaN}_x\text{As}_{1-x}$) has been discovered by Weyers, Sato, and Ando in 1992,¹ and the band-gap reduction has been demonstrated clearly for the first time. Recently, N-containing alloys such as $\text{Ga}_x\text{In}_{1-x}\text{NAs}$ have been attracting increasing interest for the next generation optical fiber communication devices to be fabricated on a GaAs substrate. That research interest is based on the anomalously large optical bowing effects. In the impurity limit, it is well known that the localized electronic states in N-doped GaP (GaP:N) have been utilized for green light-emitting devices during the past few decades. As well as the case of GaP:N, N doping in GaAs (GaAs:N) creates a series of N-related localized states.²⁻⁹ Especially, localized states within the conduction band are considered to result in significant band-structure changes in the host material.^{10,11} On the other hand, many of the N-induced bound states have been observed in spectroscopic measurements,²⁻⁹ in which discrete emission lines have been attributed to N-N pairs and N clusters. Each emission line is close to resolution-limited luminescence. Such extremely narrow bandwidth luminescence can be applicable to a new light source with a defined photon energy. Recently, it has been demonstrated that each individual bound exciton in ZnSe generates triggered single photons.¹² The concept of the single-photon emission from each individual impurity center embedded in a resonator is very promising. We can expect similar phenomena in GaAs:N, in which it is important to note that the emission wavelength matches with the infrared optical communication band. The single-photon source has to include a single emission center in a device. Therefore, the development of well-controlled N-doping techniques is prerequisite.

In this work, we have developed a site-controlled N-doping technique using molecular-beam epitaxy (MBE). The N doping has been performed in an atomically controlled way using the (3×3) -N stable surface on GaAs(001), which has succeeded in forming selected isoelectronic cen-

ters. To study detailed bound exciton states at the isoelectronic centers, we performed photoluminescence (PL) spectroscopy.

II. SITE-CONTROLLED DOPING OF NITROGEN AND OPTICAL MEASUREMENTS

N atomic-layer doped GaAs has been grown on an undoped GaAs(001) substrate by MBE. Before doping N, a 380-nm thick GaAs-buffer layer was grown at 565 °C. The As flux was 3.0×10^{-6} Torr. We used active nitrogen species created in a radio-frequency plasma source from ultrapure N_2 gas. The gas-flow rate was 1.0 ccm. The N doping has been performed at 570 °C in an atomically controlled way using a N-stabilized (3×3) surface on the GaAs(001)-buffer layer. During the N doping, the As shutter was not closed. After that, a 50-nm thick GaAs capping layer was grown.

To perform site-controlled doping, we use the N-stabilized ordered surface. The reflection high energy electron diffraction (RHEED) patterns observed before and after nitridation are shown in Fig. 1. The electron-beam incident azimuth in these pictures is $[\bar{1}10]$. Nitridation on GaAs(001) surfaces is known to form the (3×3) reconstructed structure. We performed nitridation onto various (2×4) -GaAs(001) surfaces prepared at different temperatures. From these experiments, the $(2 \times 4)\alpha$ surface is found to give us the most stable (3×3) structure. At this moment, the exact (3×3) structure is not clear. Although there are no reports about the (3×3) structure on the $(2 \times 4)\alpha$ surface, the N-stabilized surface on the As-rich $(2 \times 4)\beta_2$ of GaAs(001) has been investigated by high-resolution scanning tunneling microscope images.^{13,14} Here, we focus on a model proposed by Gwo *et al.* (Ref. 13) for the (3×3) surface that appeared on the $(2 \times 4)\beta_2$ surface. The proposed model is illustrated in Fig. 1(c). They have suggested $[\bar{1}10]$ -N dimmers and a regular array of missing N rows in both $[\bar{1}10]$ and $[110]$. Xue *et al.* has claimed that the N-dimer formation is unstable on the reconstructed GaN(001) surface because of a very small covalent radius of a N atom compared to that for Ga.¹⁴ Thus, at this moment the detailed dimmer structure is not clear. The N

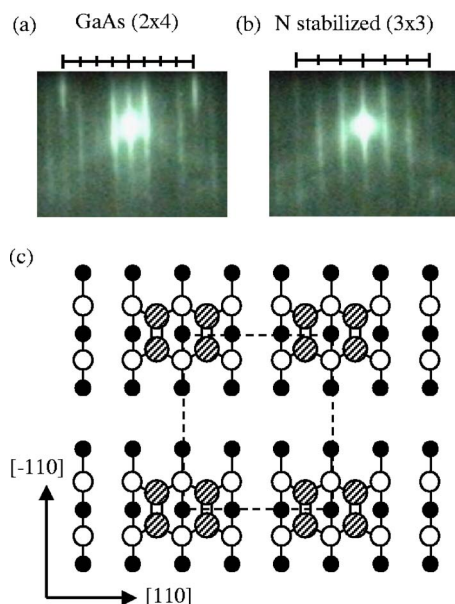


FIG. 1. (Color online) RHEED patterns before (a) and after (b) nitridation. Proposed (3×3) surface model is illustrated in (c). The dashed lines indicate the primitive unit cell. Open and closed circles represent Ga and As atoms, respectively. Nitrogen atoms represented by hatched circles form $[\bar{1}10]$ dimmers.

concentration of the proposed (3×3) surface model is $5.6 \times 10^{14} \text{ cm}^{-2}$. Since our nitridation has been performed on the $(2 \times 4)\alpha$ surface prepared at the higher temperature, the concentration is expected to be diluted. The nitridation at the raised temperature enhances step-etching caused by the N atoms,¹⁴ which creates a dilute N-doped surface maintaining the long-range ordered structure. When capping the N-stabilized surface, we have confirmed layer-by-layer growth. The RHEED specular-beam intensity shows a clear oscillation representing the monolayer growth during the capping layer deposition, which suggests an atomically controlled doping of N atoms. The sheet concentration of the doped N analyzed by secondary ion mass spectroscopy is about $3 \times 10^{12} \text{ cm}^{-2}$.

Detailed PL measurements on the GaAs:N sample were carried out to study exciton states of the N-isoelectronic centers. The excitation has been done by the 488-nm line of an argon-ion laser. The samples were mounted on a copper block cooled at 4 K in a closed cycle cryostat. On the other hand, in magneto-PL measurements, the samples were set in a superconducting-magnet system at 2.5 K in a He-gas-flow cooling system. PL was dispersed in a 550-mm single monochromator and detected by a liquid-nitrogen cooled Si-charge coupled device array. The resolution limit of our system is 0.1 meV.

III. RESULTS AND DISCUSSION

A. PL properties of GaAs:N

Figure 2 is a typical PL spectrum of the GaAs:N sample at 4 K. The excitation density was about 200 W/cm^2 . The spectrum shows only two major narrow lines at 1.4431 and

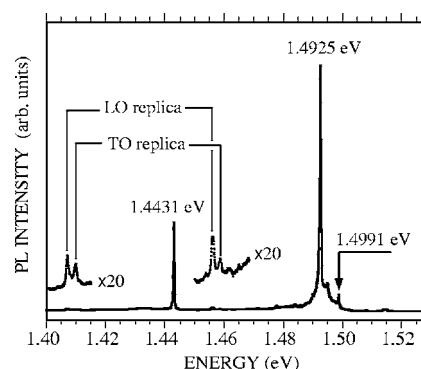


FIG. 2. PL spectrum of GaAs:N at 4 K.

1.4925 eV with the spectral linewidth of 0.5 meV, respectively. Near the 1.4925-eV line, several weak signals are observed. The number of the observed PL lines is limited extremely as compared to previous results on PL coming from N-related centers formed in GaAs:N.²⁻⁹ The PL-peak positions of the two lines are quite similar to characteristic signals of isoelectronic traps in GaAs:(In,N),¹⁵ although the PL intensity of our atomic-layer doped GaAs is remarkably high. The comparison between GaAs:N and GaAs:(In,N) is discussed in Sec. III E. It is well known that the discrete PL properties strongly depend on the N-doping level.⁹ Many literatures studied for GaAs:N with various N-doping levels have lead to controversial assignments for N-related bound states.²⁻⁹ The N-doping performed here selects a few structures of the large number of the N aggregates expected to be a small fraction of the total nitrogen concentration in a uniformly doped GaAs:N. That can limit the number of the PL lines. As magnified in the inset, the PL line is accompanied by LO- and TO-phonon replicas. That is a characteristic feature of an isoelectronic center, and, therefore, means that the exciton is localized at the luminescence center.

The PL intensity of these lines is reduced rapidly with raising temperatures. Only a slight decrease of the intensity within 10% is observed in the low-temperature region up to about 10 K. A marked decrease of the PL intensity is observed at temperatures greater than 20 K.

To investigate the spatial distribution of the bound exciton wave function, we have carried out linearly polarized PL measurements for the 1.4431- and 1.4925-eV lines. Open and closed circles in Fig. 3 indicate polarized PL intensities as a function of the polarization angle. The polarization angle is defined from $[\bar{1}10]$. Both the PL intensity profiles show a clear sinuous behavior, in which the intensity maxima correspond to the $[110]$ polarization. This result demonstrates an anisotropic distribution of the N-related bound states along the $[110]$ direction. Because of the anisotropic behavior, the lowest (1.4431 eV) and second lowest (1.4925 eV) transitions can be attributed to the N aggregates such as N-N pairs ordered along the $[110]$ direction. The point group of the $[110]$ -ordered defect is low symmetry C_{2v} , consisting of a twofold axis along the $[001]$ and a reflection plane.

Recently, Kent and Zunger reported theoretically calculated electronic structures of GaAs:N.¹⁶ Their calculated energy levels for N-N pairs in GaAs are a *nonmonotonic* func-

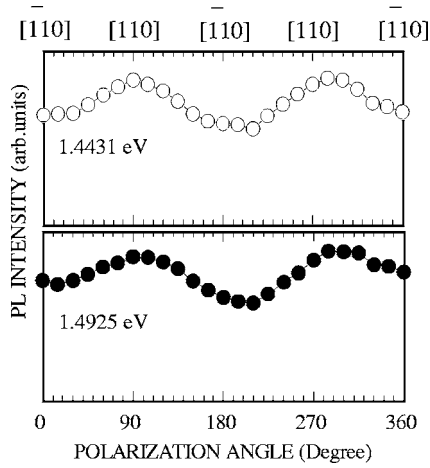


FIG. 3. Linearly polarized PL intensity of GaAs:N as a function of a polarization angle.

tion of pair separation. It is noted that only the first $[1,1,0]$ and fourth $[2,2,0]$ nearest neighbor pairs create levels having strong Γ character within the band gap. The calculated energy levels for the first and fourth nearest neighbor pairs are close to each other, whose energies seem to agree moderately with the 1.4431-eV line. Furthermore, the $[110]$ anisotropy of these pairs is consistent with the polarized PL result shown in Fig. 3. Since the (3×3) -ordered structure consists of units of the first nearest neighbor pairs, the 1.4431-eV line can be assigned to be due to the first nearest neighbor pair. There are in total twelve undistinguishable directions to the first nearest neighbor pairs in the zincblende structure. Four of them are in the (001) plane. It is easy to understand why the atomic-layer doping in the (001) plane selects the four pairs. Furthermore, two of the four pairs are ordered along $[110]$. The selection of the $[110]$ pairs from the four, however, is not clear. A couple of the $[\bar{1}10]$ -N dimmers as shown in Fig. 1(c) is difficult to explain the preferential $[110]$ pairs, but the observed preferential pair may suggest formation of N-As dimmers instead of the N-N dimmers nevertheless. On the other hand, they have predicted that defect states of Ga-centered clusters $\text{Ga}(\text{As}_{4-p}\text{N}_p)$ with $p=3$ and 4 and $[110]$ chains longer than the N-N pair appear in an energy range deeper than the $[1,1,0]$ N-N pair level. However, we did not observe significant PL signals related to such structures below 1.4431 eV.

In contrast to the assignment of the 1.4431-eV line, it is difficult to find the origin of the shallow level at 1.4925 eV. Actually, the simple N-N pair structures considered in Ref. 16 fail to explain a large number of experimentally observed shallow levels²⁻⁹ located below the conduction-band edge. The $[110]$ -polarization anisotropy of the 1.4925-eV line suggests that the aggregate also consists of a pair structure along $[110]$. As pointed out in Ref. 16, that can be attributed to short chains of N atoms or a pair with much longer distance. That was confirmed by changing the doping concentration. Here, the doping concentration was controlled by the nitridation time. With increasing the concentration, the emission intensity of the 1.4925-eV line has been found to be reduced

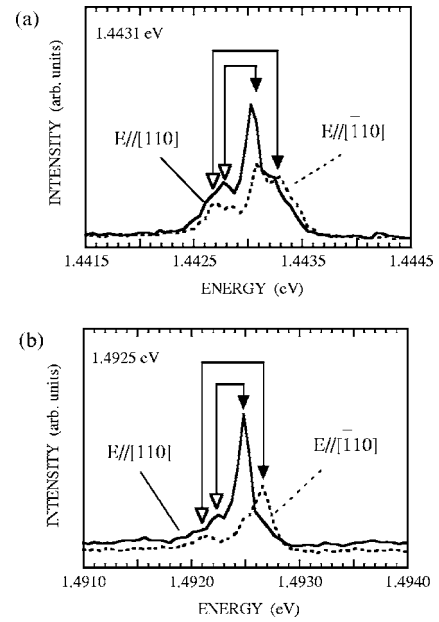


FIG. 4. Polarized PL spectra for 1.4431- and 1.4925-eV lines of GaAs:N measured at 2.5 K.

rapidly, because the higher concentration gives rise to a shorter mean distance of N pairs.

B. Fine structure of the 1.4431- and 1.4925-eV lines

Figure 4 shows highly resolved polarized PL spectra measured at 2.5 K for the 1.4431- and 1.4925-eV lines. The $[110]$ ($[\bar{1}10]$) polarization corresponds to the direction showing the PL-intensity maxima (minima) in Fig. 3. The spectrum is found to show fine structures. Since the He-gas-flow cooling system can achieve a lower sample temperature than that of the closed cycle cryostat, such fine structures are considered to be resolved even at the same excitation density (200 W/cm^2). The structure consists of four signals, which can be selected by the polarization. Each spectral linewidth is close to our resolution limit (0.1 meV). The $[110]$ ($[\bar{1}10]$) polarization selects the inner (outer) pair. The selection is very clear for the 1.4925-eV line in contrast to the 1.4431-eV line. Since the pair separation is larger for the higher-energy state, the 1.4925-eV line is considered to have the pronounced anisotropy as compared with the 1.4431-eV line.

C. Magneto PL of GaAs:N

Magneto PL is a powerful technique to reveal the structure of the bound exciton state. The typical Zeeman diagram for the 1.4925-eV line of GaAs:N is shown in Fig. 5. The measurement was carried out at 2.5 K. Here, the polarization is parallel to the $[110]$ direction. The magnetic field was applied along the $[\bar{1}10]$ direction and the magnitude was changed up to 7 T. It is noted that the Zeeman shift demonstrates splitting into three levels, which is a direct evidence that the bound exciton is triplet. The bound exciton state is formed from the bound-hole and bound-electron states, which is coupled by the Coulombic exchange interaction.

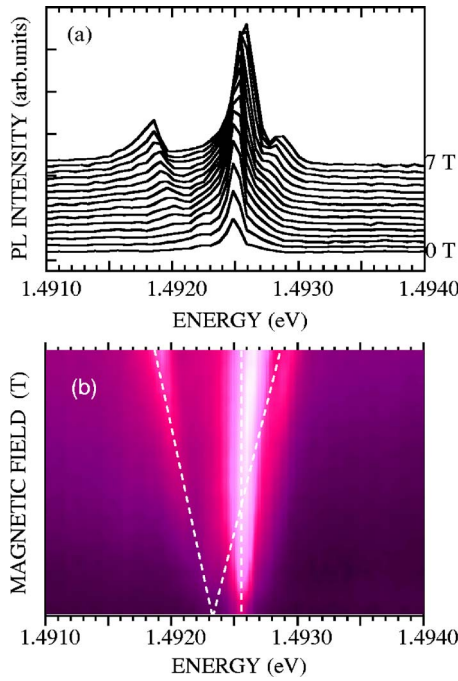


FIG. 5. (Color online) Typical Zeeman diagram for the $[\bar{1}10]$ -polarized 1.4925-eV line of GaAs:N in magnetic fields parallel to $[\bar{1}10]$. (a) PL spectra measured at various magnetic fields and (b) a counter plot of the PL intensity as a function of magnetic field.

The coupling produces fine-structure splitting. The exciton-spin wave functions are given by three symmetrical $s=1$ spin wave functions and one antisymmetrical $s=0$ spin wave function. The spin triplet exciton (dark exciton) is a dipole-forbidden state, while the spin-singlet exciton (bright exciton) is a dipole-allowed state. Luminescence from such spin triplets has been reported for highly localized bound excitons in semiconductors.¹⁷ The observed phonon replicas in the spectrum supports that the exciton is significantly localized at the luminescence center.

Moreover, the zero-field splitting of the triplet state is observed clearly. The Zeeman diagram ignoring level coupling is drawn by dashed lines in Fig. 5(b). The spin projections of the triplet exciton are 0 and ± 1 . Closed and open arrows in Fig. 4 indicate the zero-field splitting into the states with the projections of $|0\rangle$ and $|\pm 1\rangle$, respectively. The zero-field splitting is caused by the defect anisotropy. The smaller (larger) zero-field splitting of the inner (outer) pair indicates a smaller (larger) anisotropy of the defect. The two sets of the zero-field splittings are considered to be caused by the removal of the orientation degeneracy of the C_{2v} defect in the cubic lattice. A defect-induced strain field with local coordinate axis X , Y , and Z can remove some of the p -state degeneracy, leading to a lowering of one of the p_x , p_y , and p_z states relative to the others.¹⁸ For $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 001 \rangle$ stresses, there are two, three, and two inequivalent classes, respectively. Since the growth has been done on the $\langle 001 \rangle$ plane, the splitting into the two classes can be attributed to the $\langle 001 \rangle$ stress, i.e., biaxial stress in the $\langle 001 \rangle$.

The energies of the triplet exciton states $|0\rangle$ and $|\pm 1\rangle$ are given by $E_0 - \Delta$ and $E_0 \pm 0.5g\mu_B B$, respectively, where g is the

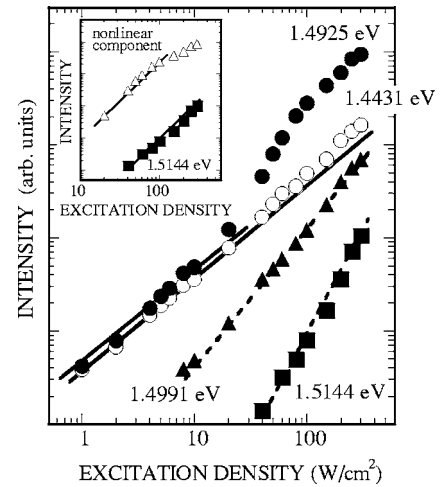


FIG. 6. PL peak intensities for the 1.4431- (open circles), 1.4925- (closed circles), and 1.4991-eV (closed triangles) lines of GaAs:N as a function of excitation density.

$[\bar{1}10]$ component of the exciton g factor, μ_B is the Bohr magneton, and Δ is the zero-field splitting. Though the Zeeman diagram for the 1.4431-eV line is complicated because of simultaneous observation of the nondegenerated states as shown in Fig. 4(a), the field evolution demonstrates the similar triplet-exciton character. The Zeeman shifts for the 1.4431- and 1.4925-eV lines are found to be slightly different. The shift for the 1.4431-eV line is larger than that of the 1.4925-eV line. The g factors estimated from the field evolution of the $|-1\rangle$ state for the 1.4431- and 1.4925-eV lines are 2.4 and 1.9, respectively. The larger g factor of the deeper bound state represents the stronger exciton localization.

D. Excitation-power dependence of PL from GaAs:N

The excitation-power dependence of the PL intensity is shown in Fig. 6. Open and closed circles indicate intensities of the 1.4431- and 1.4925-eV PL lines, respectively. The solid lines guide the linear power dependence. We found nonlinear responses around 30 W/cm² for both the 1.4431- and 1.4925-eV lines, which seems to be a jump in intensity. The jump is remarkable for the 1.4925-eV line, while the jump of the 1.4431-eV line is quite small. Figure 7 displays a logarithmic plot of PL spectra measured at 10, 20, 50, 100, and 200 W/cm². The jump in intensity occurs in this illumination range. On increasing the illumination, it is found that several signals whose intensity increases nonlinearly appear in between the 1.4925-eV PL line and the GaAs edge. Peak energies of these new signals (indicated by arrows) are 1.4991, 1.5081, and 1.5144 eV (just below the GaAs edge). This result suggests recombination ($X_i \rightarrow X_{i-1}$) of exciton complexes X_i in the confined electronic shells as demonstrated in semiconductor quantum dots.¹⁹ The isoelectronic center is considered to be an ultimate quantum dot. For example, the power dependence of the 1.4991-eV line is shown in Fig. 6 by closed triangles, which are fitted by the dashed

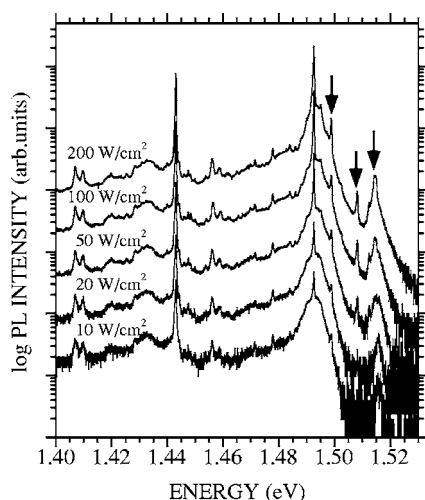


FIG. 7. Logarithmic plot of PL spectra measured at 10, 20, 50, 100, and 200 W/cm².

line with a slope of 1.5. The different power dependences exclude a model with common initial states for the different observed transitions. The slope implies that the 1.4991-eV line may be a biexciton (excitonic helium) recombination. This state is considered to be associated with the same binding center as the 1.4925-eV exciton, because the 1.4431-eV line is far from 1.4991 eV. The lowest excitonic state can be occupied by up to two excitons. Therefore, the luminescence from the higher excited state could be observed, when multiple excitons are simultaneously created in an artificial atom. The excitation power dependences of the 1.5081- and 1.5144-eV lines yield slopes of about 1.5 and 2.0, respectively. Solid squares in Fig. 6 indicate the results for the 1.5144-eV line. The energy levels of the excitonic artificial atom depend on the illumination.¹⁹ It is noted that the appearance of the jump in intensity coincides with occupation of the confined electronic shells. If excitons can transfer, in particular, from the GaAs edge by way of a shell state determined at an excitation occupation, another nonlinear excitation channel opens up. Then the following cascade annihilation of the excitons enhances the emission intensity of the ground-state PL at 1.4925 eV. We compare the nonlinear component in the 1.4925-eV signal with the nonlinear power dependence of the 1.5144-eV line which appears beneath the GaAs edge. The comparison is shown in the inset of Fig. 6. The nonlinear component in the 1.4925-eV signal was obtained by subtracting off the linear component from the power dependence above the onset. The nonlinear power dependence of the new peaks and of the 1.4925-eV line above the jump is almost the same. Then the cascade and relaxation of these new peaks are responsible for the sudden jump in intensity of the 1.4925-eV line. To understand the excitation power dependence completely, we need further detailed spectroscopic approaches.

E. Comparison of PL spectra from GaAs:N and In_xGa_{1-x}As:N

Next, we compare the above-mentioned PL features of GaAs:N with results for In_xGa_{1-x}As:N. The sample

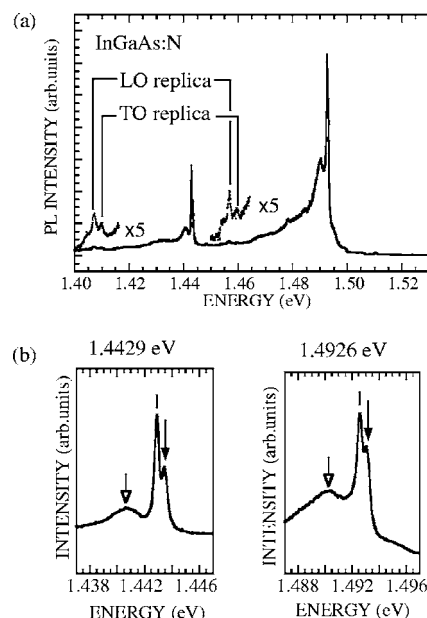


FIG. 8. (a) PL spectrum of In_xGa_{1-x}As:N. (b) Magnified spectra of 1.4431- and 1.4925-eV lines. Sample temperature is 4 K.

growth of In_xGa_{1-x}As:N has been performed by the same procedure. The atomically doped layer was sandwiched in between a 90-nm In_xGa_{1-x}As layer; InGaAs(90 nm)/N/InGaAs(90 nm). The InAs-mole fraction is 0.023. Finally, a 50-nm thick GaAs caps the InGaAs(90 nm)/N/InGaAs(90 nm) layer. Figure 8(a) shows a PL spectrum of In_xGa_{1-x}As:N. The obtained spectrum is quite similar to the result of GaAs:N except for fine structures as magnified in Fig. 8(b). The main PL-peak energies (indicated by vertical bars) of In_xGa_{1-x}As:N are at 1.4429 and 1.4926 eV, which almost coincide with those of GaAs:N shown in Fig. 2. The In incorporation causes new sharp lines indicated by solid arrows. The relative PL intensity of the new line to the main peak indicated by the bar has been found to be increased with increasing the excitation light intensity and raising the sample temperature. On the other hand, a broad side-band structure indicated by open arrows appears at the lower energy side. The broad side-band structure can be attributed to quasilocalized phonon modes.¹⁷ Since the side-band structure is not observed in GaAs:N (see Fig. 2), the structure is induced by the In atoms. Therefore, In atoms are considered to tend to locate close to the N pairs.

Quite similar results have been reported for GaAs:(In,N) by Intartaglia *et al.*¹⁵ They have also observed two major PL lines from isoelectronic centers. The peak position coincides with our results. However, they have never observed the signal near ~1.443 eV in GaAs:(In,N) with low In concentrations less than 10¹⁷ cm⁻³, and, thereby, the signal has been attributed to the radiative recombination of excitons bound to N clusters *perturbed by one In atom*. Obviously that result is different from ours. The 1.4431-eV PL line is remarkable even in GaAs:N. Therefore, now we conclude that the 1.4431- and 1.4925-eV lines are characteristic of GaAs:N itself. Intartaglia *et al.* have assigned the main peak indicated by the vertical bar and the newly appeared peak indicated by

the solid arrow in Fig. 8(b) to the $J=2$ and $J=1$ exciton transitions, respectively.¹⁵ As mentioned above, we attribute the 1.4431- and 1.4925-eV lines of GaAs:N to the triplet exciton transition. That assignment is consistent with the results derived by Intartaglia *et al.*¹⁵ However, the assignment for the newly appeared peak indicated by the solid arrow is different. Two important questions are remaining; (1) why is the peak energy of the main PL line independent of the In doping? and (2) why does the In doping create the new emission line indicated by the solid arrow? Although it is considered that the combination of In and N reduces the amount of elastic strain, obvious influence of the strain on the peak position is not observed. This puzzle gives rise to another scenario for the split PL lines in $\text{In}_x\text{Ga}_{1-x}\text{As:N}$. If we assume that the main (vertical bar) and new (solid arrow) lines come from the radiative recombination of excitons bound to N pairs unperturbed and perturbed by In atoms, respectively, the questions (1) and (2) seem to be solved.

IV. CONCLUSIONS

We performed site-controlled doping of N into GaAs using the N-stabilized (3×3) ordered surface. The bound exciton states of isoelectronic centers show two remarkable

narrow bandwidth PL lines. The polarized PL shows that the luminescence centers consist of N-pairs bonding along the [110] direction. Magneto PL is used to reveal the structure of the exciton ground states. The Zeeman shift gives us a direct evidence of the triplet state. With increasing illumination, we found a significant intensity jump for the 1.4925-eV line. Furthermore, it is found that several signals whose intensity increases nonlinearly appear in between the 1.4925-eV PL line and the GaAs edge. These results are supposed to be due to a formation of exciton complexes. We compared PL features of GaAs:N with results for $\text{In}_x\text{Ga}_{1-x}\text{As:N}$. The In incorporation causes a new sharp line as well as a broad side-band structure owing to a quasilocalized phonon mode. We conclude that the 1.4431- and 1.4925-eV lines are found to be characteristic of GaAs:N itself.

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